

# NUCLEAR SPIN RELAXATION AND INCOMMENSURATE MAGNETISM IN DOPED CUPRATES

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**Abstract** Existing data on  $^{63}\text{Cu}$ -nuclear spin relaxation reveal two independent relaxation processes: the one that is temperature independent we link to incommensurate peaks seen by neutrons, while the "universal" temperature dependent contribution coincides with  $1/^{63}T_1(T)$  for two-chain YBCO 124. We argue that this new result substitutes for a "pseudogap" regime in a broad class of high- $T_c$  cuprates and stems from the 1st order phase transition that starts well above the superconductivity  $T_c$  but becomes frustrated because of broken electroneutrality in the  $\text{CuO}_2$  plane.

**Keywords:** superconductivity, pseudogap, magnetic properties, NMR

## 1. Introduction

One of the most intriguing normal properties of the high- $T_c$  ( $\text{HT}_c$ ) cuprates is the so called "pseudogap" (PG) phenomenon. It is commonly presented in the  $(T, x)$  plane as a line that starts from rather high temperatures (at small  $x$ ) and reaches the superconductivity (SC)  $T_c$  "dome" below at or above optimal  $x \sim 0.16$ . In a broad sense  $x$  means the hole concentration in the  $\text{CuO}_2$ - plane, but more often than not one refers to properties of the Sr-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . The PG feature was seen in numerous experiments (NMR, tunneling spectra, resistivity etc.;

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see for example reviews [1, 2]). It has been stressed [3] that the PG temperature is not defined unambiguously.

A widespread view is that the feature comes from some crossover in the electronic density of states (DOS). The main result of the present paper is that after a proper re-arrangement of the experimental data no PG feature exists in the  $^{63}\text{Cu}$  nuclear spin relaxation time behaviour. Instead, the data show two independent parallel relaxation mechanisms: a temperature independent one that we attribute to stripes caused by the presence of external dopants and an "universal" temperature dependent term which turns out to be exactly the same as in the stoichiometric compound YBCO 124.

## 2. The experimental results and discussion

We attempt below to put the results in the context of a phase separation [4]. The decomposition of  $1/^{63}T_1(T, x)$  into two terms, as it will be discussed below in more details, manifests itself in a broad temperature interval above  $T_c$ . It is limited from above by a  $T^*$  that depends on the concentration,  $x$ . We consider  $T^*$  defined in this way as a temperature of a 1st order phase transition, which, however, cannot complete itself in spatial coexistence of two phases because of the electroneutrality condition [5]. It was already argued in [4] that such a frustrated 1st order phase transition may actually bear a dynamical character. The fact that a single resonant frequency for the  $^{63}\text{Cu}$  nuclear spin is observed in the NMR experiments, confirms this suggestion. Although in what follows, we use the notions of the lattice model [4, 5], even purely electronic models [6–9] for cuprates may reveal a tendency to phase separation.

The basic assumption in [4, 5] are the following. At large enough doping holes move between coppers and oxygens. Spins in the system are  $d^9$ -holes trapped to the Cu-sites at the expense of local lattice distortions. Elastic attractive interactions between these distortions give rise to a lattice driven frustrated transition below some  $T^*$ . Exchange interactions, as in the parent  $\text{La}_2\text{CuO}_4$ , tend to organize the Cu-spins in the antiferromagnetic (AF) sub-phases. Excess charge of the dopants' ions in AF regions must be compensated by accumulation of holes in "metallic" regions.

We now turn to experimental data. In what follows we address only  $1/^{63}T_1$  behaviour because for cuprates AF fluctuations prevail over the Korringa mechanisms.

In Fig. 1a we collected data on  $1/^{63}T_1$  in LSCO from [10]. Note the following: 1) according to [11]  $1/^{63}T_1(T)$  at higher temperatures tends to  $2.7 \text{ msec}^{-1}$  for all Sr concentrations, in spite of considerable spread

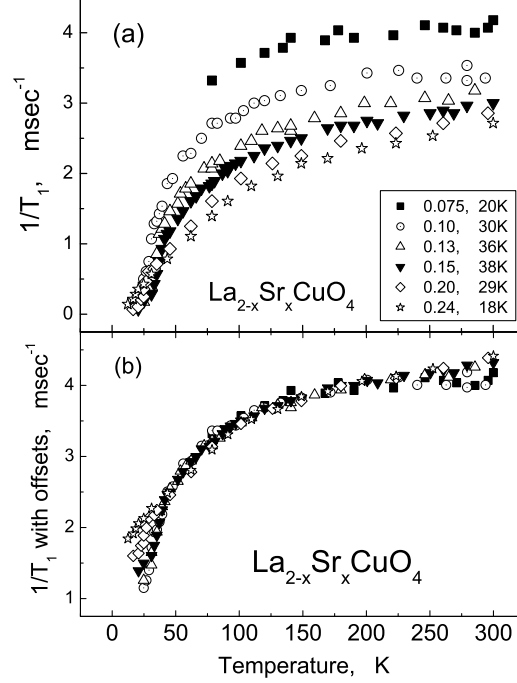


Figure 1. The temperature dependence of  $1/^{63}\text{T}_1(x)$  for LSCO: a) the plots for different  $x$  and  $T_c$  (see inset) are taken from [10], at higher temperatures all of them converge to the same value of 2.7 msec<sup>-1</sup> [11]; b) the same dependencies collapsing to the single curve after the corresponding vertical offsets.

seen in Fig. 1a. Beginning of deviation from that value could serve us as a definition of  $T^*(x)$ ; 2) note that dissipation  $1/^{63}\text{T}_1$  monotonically decreases from small  $x$  to 0.24; 3) after an appropriate *vertical* offset all curves in Fig. 1a collapse onto the  $T$  dependence of  $1/^{63}\text{T}_1$  for the “optimal”  $x = 0.15$  above 50 K (Fig. 1b). We have checked that last tendency works well for YBCO (6.5) doped with Ca i.e., the data for different  $z$  in  $\text{Y}_{1-z}\text{Ca}_z\text{Ba}_2\text{Cu}_3\text{O}_{6.5}$  [12] may be put on the top of each other after proper offsets.

This prompts us to verify whether same “off-settings” of the  $1/^{63}\text{T}_1$  data apply to a broader group of materials. The stoichiometric  $\text{YBa}_2\text{Cu}_4\text{O}_8$  possesses no structural or defect disorder and we adjust all data to the  $1/^{63}\text{T}_1$  behaviour for this material [3]. Fig. 2 shows that after a vertical shift in  $1/^{63}\text{T}_1$  all the materials indeed follow the same “universal” temperature dependence above their  $T_c$  and below 300 K.

In other words, in this temperature range the nuclear spin relaxation in these cuprates is a sum of contributions from two parallel processes:

$$1/^{63}T_1 = 1/^{63}\overline{T}_1(x) + 1/^{63}\widetilde{T}_1(T) \quad (1)$$

In eq. (1)  $1/^{63}\overline{T}_1(x)$  depends on a material and a degree of disorder ( $x$ ), but does not depend on temperature, while  $1/^{63}\widetilde{T}_1(T)$ , depends only on temperature, is the same for all these compounds and coincides with the  $1/^{63}T_1$  for the two chains YBCO 124 above its  $T_c=62$  K.

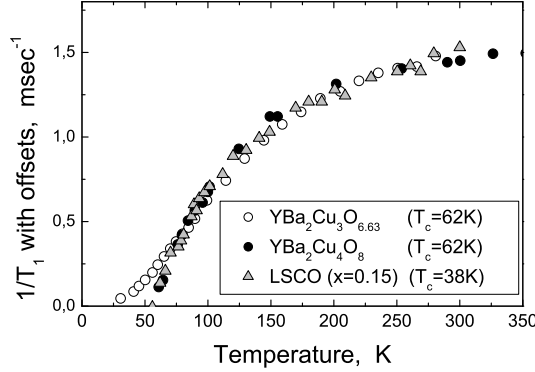


Figure 2. The temperature dependence of  $1/^{63}T_1$  for different compounds: the  $1/^{63}T_1$  for YBCO (123) [13] overlayed with that for LSCO [10] and YBCO (124) [3].

Thus, to some surprise the only "pseudogap" feature in the NMR data that may be discerned in Fig.2 is the one for YBCO 124: a change in the temperature regime between 130 and 180 K. It would be tempting to take again this feature as a mark of the PS regime taking place now in the stoichiometric material where doping most definitely comes about as a spill-over of carriers from the CuO-chains into CuO<sub>2</sub> planes. It is also natural to think that the number of the transferred carriers is not small: actually the low temperature Hall effect measurements [15–17] show a rapid increase in the number of carriers (i.e. Fermi surface size) up to one hole per unit cell even in the single layer material like LSCO, at the optimal doping  $x \sim 0.15$ . Recall, however, that little is known for the "homogeneous" phase (i.e. above  $T^*(x)$ ). Properties of both YBCO 124 and the optimally doped LSCO (see [1] for review) are unusual and best described in a very broad temperature interval in terms of the "marginal" Fermi liquid [18]. We have not found a reliable experiment to define  $T^*$  for these compounds and therefore leave the origin of the  $1/^{63}\widetilde{T}_1(T)$ -term for further discussions.

The decomposition (1) into two parallel dissipation processes show that usual definitions of  $T^*$  [19] have no grounds. In Fig.1a the LSCO data with  $x < 0.15$  are spread even above 250 K. As a rough estimate for  $T^*$ , it is much higher than the SC onset temperature.

Fig. 3 presents the dependence on  $x$  for  $1/^{63}\overline{T}_1$  in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . The inset provides the "offsets" (i.e.  $1/^{63}\overline{T}_1$  terms) for other materials. We return to discussion of Fig.3 later.

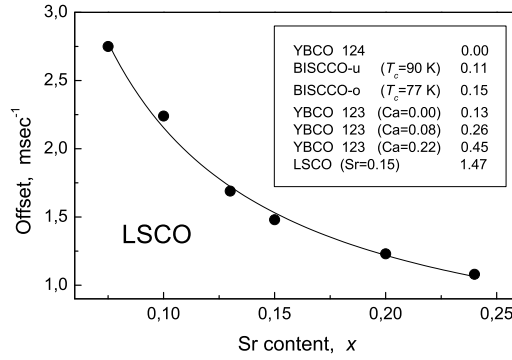


Figure 3. The offset  $1/^{63}\overline{T}_1(x)$  vs Sr content  $x$  for LSCO (relative to that for YBCO 124), line is a guide for eyes. Inset: the offsets for some other compounds (data for underdoped (u) and overdoped (o) BISCCO 2212 deduced from [14]; to compare BISCCO with LSCO and YBCO materials the hyperfine constants have to be properly adjusted).

The observation that is central for the following is that in all the materials with non-zero  $1/^{63}\overline{T}_1$  (see the inset in Fig.3) incommensurate (IC) peaks have been observed in neutron scattering [20]. Peaks are close to the  $[\pi, \pi]$  - point: at  $[\pi(1 \pm \delta), \pi]$  and  $[\pi, \pi(1 \pm \delta)]$  [21]. We will now look for the connection between these two phenomena.

We first make an attempt to agree on a semi-quantitative level the observed IC magnetic peaks in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with the values of the first term in eq. (1). We concentrate on  $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$  for which the most detailed data are available [22].

With the notation from [23]

$$1/T_1 = \frac{k_B T}{2\mu_B^2 \hbar^2 \omega} \sum_i F(Q_i) \int \frac{d^2 q}{(2\pi)^2} \chi''(q, \omega \rightarrow 0) \quad (2)$$

where  $Q_i$  stands for one peak, hyperfine "tensor"  $F(Q) = \{A_\perp + 2B[\cos(Q_x) + \cos(Q_y)]\}^2$  and for  $\chi''(q, \omega \rightarrow 0)$  we take near single peak,

say  $[\pi(1 - \delta), \pi]$

$$\chi''(q, \omega) = \frac{\chi''_{peak}(T)\omega}{[1 + (x\xi_x)^2 + (y\xi_y)^2]^2} \quad (3)$$

where  $(x, y) = (q_x - \pi(1 - \delta); q_y - \pi)$  and  $\xi_x$  and  $\xi_y$  are the correlation lengths in the two proper directions. After integration the contribution from stripes with  $q$  along the  $x$ -direction is

$$1/^{63}T_1 = \frac{k_B T}{\pi \mu_B^2 \hbar \xi_x \xi_y} \{A_{\perp} - 2B[\cos(\pi\delta) + 1]\}^2 \chi''_{peak} \quad (4)$$

Experimentally [22]  $\chi''_{peak}(T) \propto T^{-2}$  and for  $x = 0.14$   $\delta = 0.245 \sim 1/4$ . Assuming the  $T^{-1}$  dependence [22] only for the one of  $\xi$ 's,  $\xi_x$  and using for  $A_{\perp}$  and  $B$  the known values [23] one obtains:  $1/^{63}T_1 = (4/\xi_y)$  msec $^{-1}$ . With the AF correlation length  $\xi_y \sim 4$  this is the correct order of magnitude.

The descending dependence of the offset (Fig.3) agrees qualitatively with the behavior of  $\delta(x)$  [24] in eq.(4). For a quantitative description one need to know the  $x$ -dependence for  $\chi''_{peak}(T)$ . Such data in the absolute units are absent yet except [22]. Another fact that may underlie this behaviour is that with the  $x$ -increase buckling in the CuO $_2$ -planes is known to decrease diminishing pinning effects and making the local symmetry of the CuO $_2$ -unit same as in other materials from the class with small offset in Fig.3. Also, the system grows more metallic with a high holes' content [15–17].

Next comes the question concerning the origin and the role played by IC peaks and the physics of fluctuations related to them.

Discovery of IC spin fluctuations presented a challenge for explaining the NMR results for the oxygen spin relaxation times: hyperfine field "leaks" originated by the AF incommensurate fluctuations, would considerably increase the oxygen's relaxation rates, but this was not seen experimentally. Slichter (see in Ref.[25]) interpreted these contradictions in terms of "discommensurations": a periodic array of soliton-like walls separating regions with a short-range AF order. Unlike neutrons, the NMR as a local probe, does not feel the overall periodicity.

Existence of stripes looks just natural in terms of a static phase separation. At doping the system (LSCO) must screen the excess charge (Sr $^{2+}$ -ions) in AF regions. Therefore stripes of the AF ordered phase must alternate with "metallic" domain walls. The stripe arrangement by itself is nothing but an optimization of the competing Coulomb and lattice forces [7]. (The phenomena is well known in physics of surface.)

Stripes in a dynamical regime need better understanding. For instance, often the IC peaks are seen by neutrons only at low enough

temperatures or for large energy transfer at an inelastic scattering [20]. At low temperatures stripes may form a long-range order even in LSCO (at smaller  $x$ , [26]), breaking the symmetry of the ground state. A better example of the "pinned" stripe order is given by Nd (or Eu)-doped LSCO [27–30]. ( $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  reproduces all features of  $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ , including SC and same positions for IC peaks at given  $x$ .) The transition into the ordered stripe phase is driven by appearance first of the lattice/charge peaks [29]. At finite temperatures stripes could be viewed as a new type of excitations above the ordered state.

On the other hand in LSCO itself "stripes" are seen through the inelastic scattering processes for arbitrary low energy transfer even at high temperatures 100-300 K [22]. while the ordered IC ground state sets in only well below, at about  $T \sim 30\text{K}$  [26]. This example provides the argument against treating "stripes" as "excitations": at so high a temperature the underlying "long-range" IC ground state would be already melted. Therefore the two-phase description seems to be closer to reality meaning that in the dynamical regime the AF regions get coupled via Coulomb forces with the "metallic" layer. Note that with the further  $x$ -increase  $\delta(x)$  increases as well and saturates making it meaningless to speak in terms of a strictly "monolayer" wall already above  $x \sim 0.14$ , where  $\delta \approx 1/4$  [22]. (Note the difference in notations for IC peaks: ( $\delta$  from [22] equals  $2\epsilon$  from [29] equals  $2\delta$  from [24]). Fig.3 demonstrates same tendency to increase the share of the "metallic" fraction with increase of Sr-concentration:  $1/^{63}\overline{T}_1(x)$  continues to drop with  $x$  above  $1/8$ .

Coexistence of a SC and the IC AF phases at low temperatures was confirmed recently by the neutron diffraction experiments [31] for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $x = 0.10$ ) in the vortex state. (The coexistence of SC and AF formations was found also from the  $\mu\text{SR}$  spectra [32]). The way of the "coexistence" of SC and the stripe order in the same sample remains unresolved: one view treats the new stripe symmetry as a superstructure superimposed on the Fermi surface that changes the energy spectrum like any SDW/CDW can do it (e.g. [33]). Another plausible alternative would be a spatially inhomogeneous coexistence of the nonsuperconducting IC AF phase and a "metallic" phase with strong fluctuations.

### 3. Summary

We have found that in a temperature interval above  $T_c$  and below some  $T^* \sim 300\text{ K}$  the nuclear spin relaxation for a broad class of cuprates comes from two independent mechanisms: relaxation on the "stripe"-like exci-

tations that leads to a temperature independent contribution to  $1/^{63}T_1$  and an “universal” temperature dependent term. For  $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$  we obtained a correct quantitative estimate for the value of the first term. We concluded from eq.(1) and Fig.3 that ”stripes” always come about with external doping and may be pinned by structural defects. We argue that the whole pattern fits well the notion of the dynamical PS into coexisting metallic and IC magnetic phases. Experimentally, it seems that with the temperature decrease dynamical PS acquires the static character with the IC symmetry breaking for AF phase dictated by the competition between the lattice and the Coulomb forces. The form of coexistence of the IC magnetism with SC below  $T_c$  remains not understood as well as behaviour of stoichiometric cuprates.

#### 4. Acknowledgements

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